Development of Laser Isotope Separation (LIS) of ^{48}Ca for the search of neutrinoless double beta decay by CANDLES

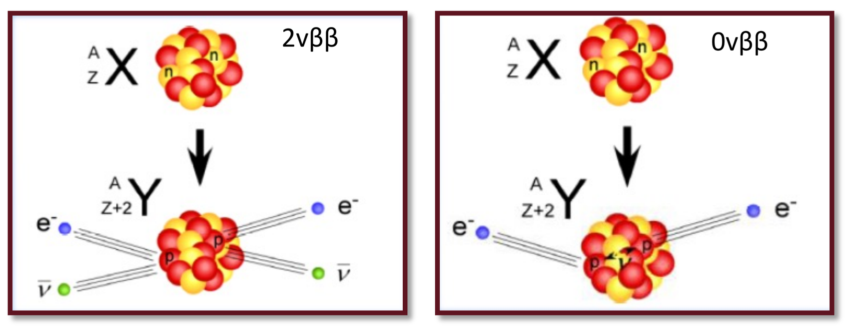
Anawat Rittirong*, LIS and CANDLES collaborations

UGRP2024, Osaka University, July 05th – 06th, 2024

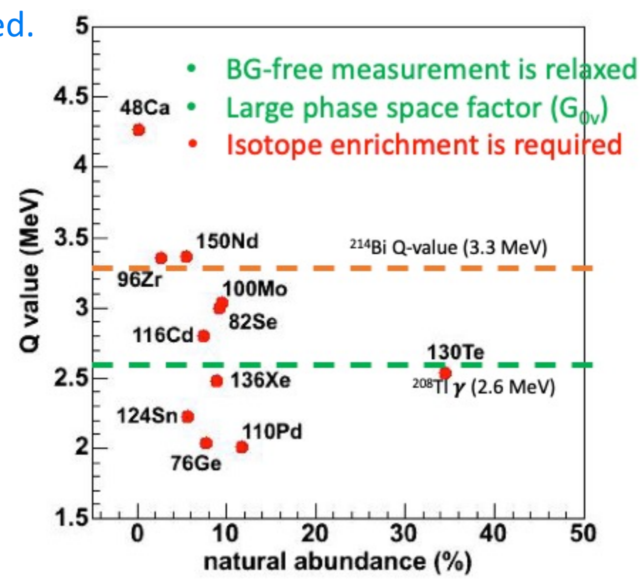
CANDLES

Candles

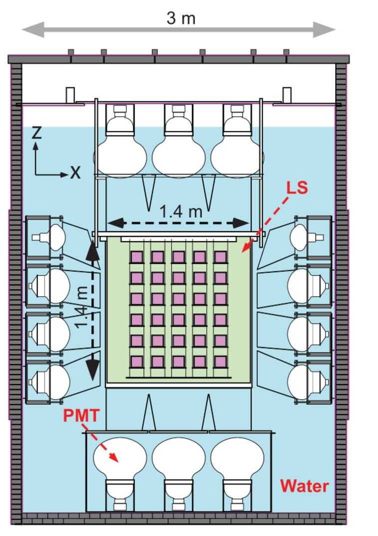
- **Purpose:** To verify the Majorana nature of neutrino via neutrinoless double beta decay (0νββ).
- **^{48}Ca** (Q-value =4.27 MeV, highest energy for 2ββ decay) is used.



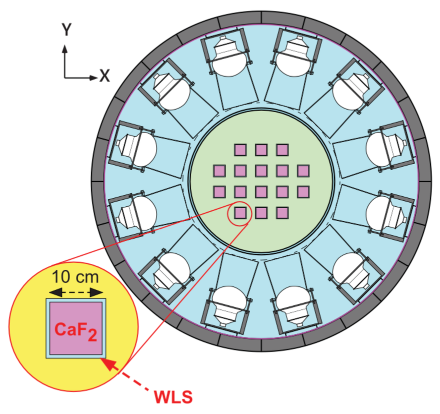
0νββ is beyond the standard model!!



https://www-conf.slac.stanford.edu/ssi/2010/Hall080610.pdf

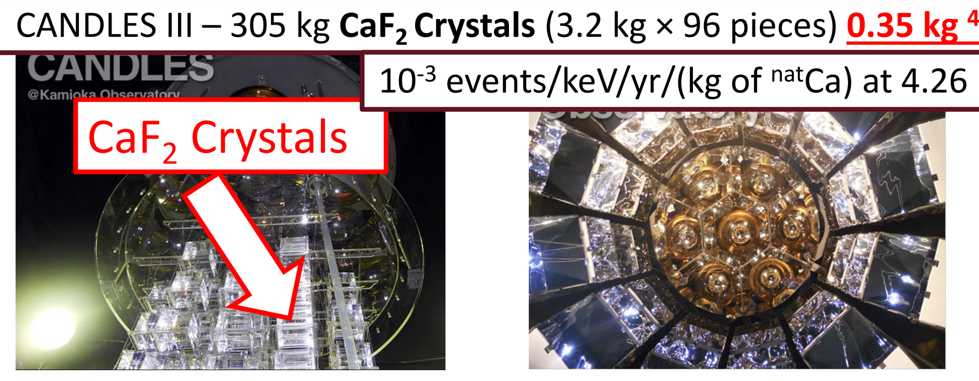


S. Ajimura, et al. *Physical Review D* 103.9 (2021): 092008.



CANDLES III – 305 kg **CaF_2 Crystals** (3.2 kg × 96 pieces) **0.35 kg ^{48}Ca**

10^{-3} events/keV/yr/(kg of ^{nat}Ca) at 4.26 MeV



Large amount of ^{48}Ca is required (tons scale)

- Regrettably, the gas centrifuge or gas diffusion techniques are not viable for the isolation of calcium due to the unavailability of the gaseous compound.

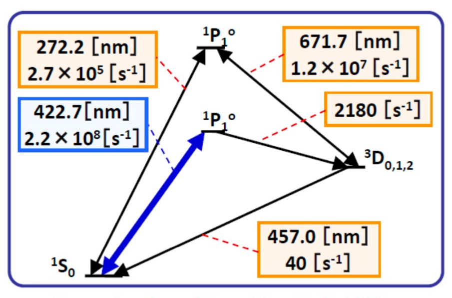
Expensive [~1,000,000 \$/g] via an electromagnetic separator

Requirement: ~0.36 – 13 kmol of ^{48}Ca ~1800 × CANDLES III

Ca Isotope	^{40}Ca	^{42}Ca	^{43}Ca	^{44}Ca	^{46}Ca	^{48}Ca
% abundance	96.94	0.65	0.135	2.09	0.004	0.187

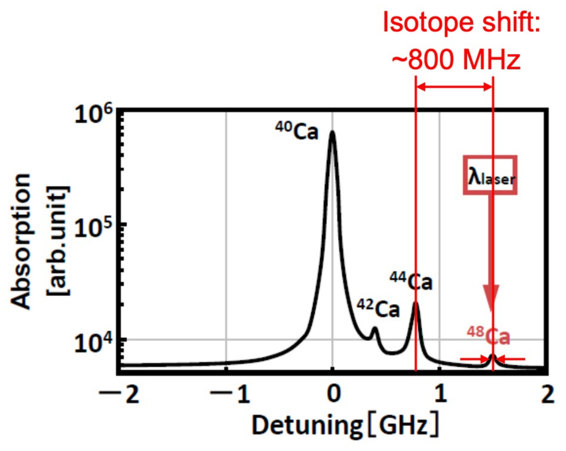
Laser Isotope Separation (LIS)

1. Proof of principle

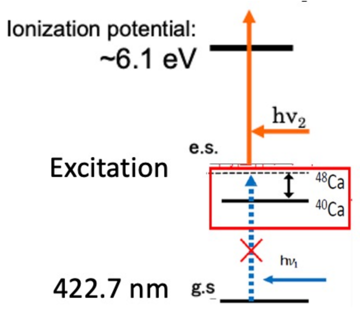


Energy levels and Transition Probabilities

Transition : $^1P_1$ - $^1S_0$
λ = 422.7 nm
τ = 4.6 ns
$\Delta\nu_{nat} = \frac{1}{2\pi\tau}$ = 34MHz (HWHM)
I_S = 59.9 mW/cm^2

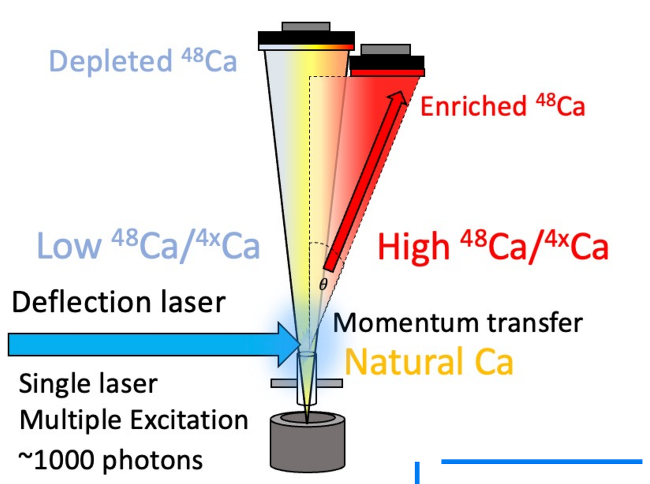


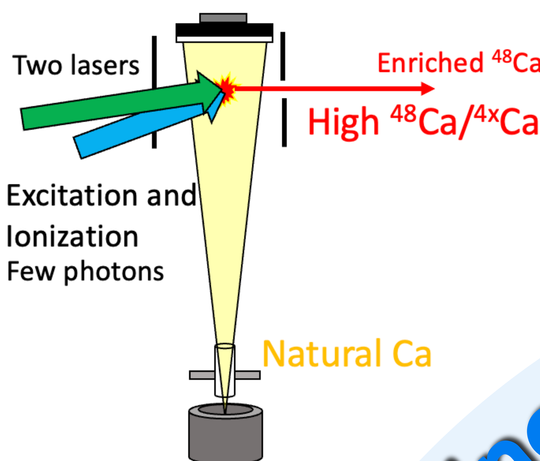
Absorption spectrum of Ca at 423nm

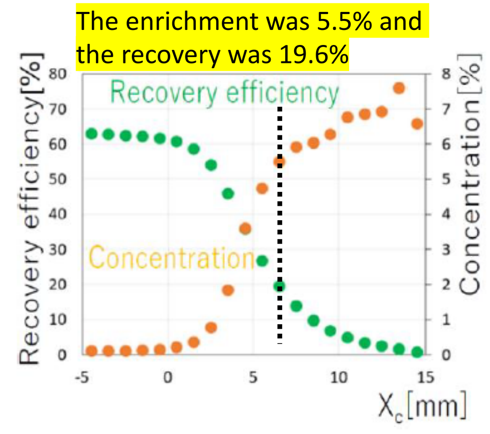


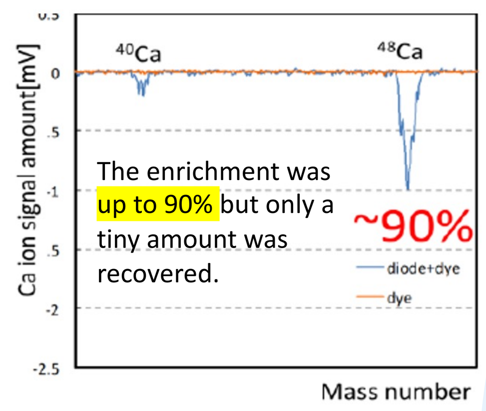
2 MHz rms → 422.792xxxxx ±0.0000006 nm

- Deflection method
- Ionization method









K Matsuoka *et al* 2020 *J. Phys.: Conf. Ser.* **1468** 012199

The DEFLECTION method is applicable for mass production.

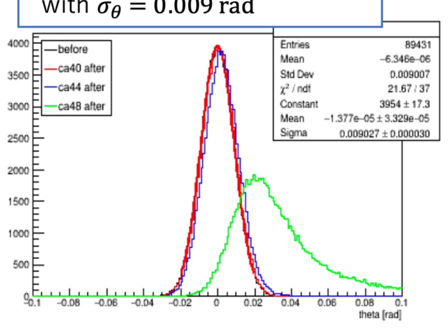
2. Atomic beam

UNIVERSITY OF FUKUI

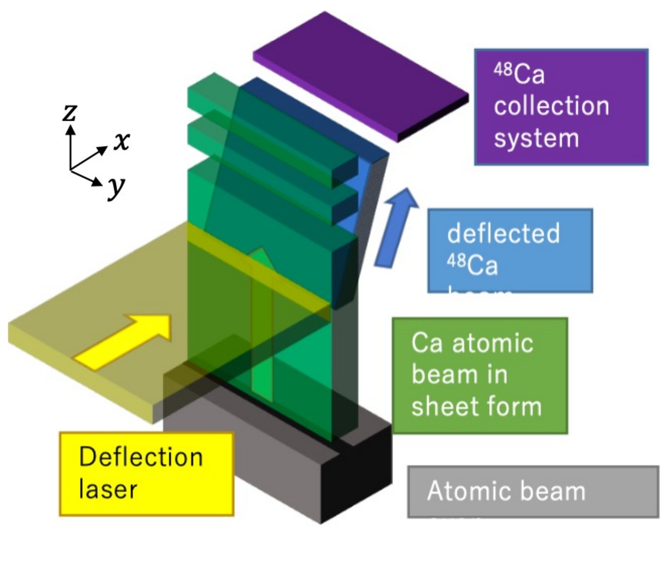
Requirements for an atomic beam system

- High intensity
  - Increase crucible temperature (600 °C)
- Well-collimated and highly efficient
  - Develop the collimator
  - Maximum allowable spread angle

Angular distribution of atoms with σ_θ = 0.009 rad



Setup simulation by GPS
- Gaussian beam at 873 K
- Laser power: 85 mW



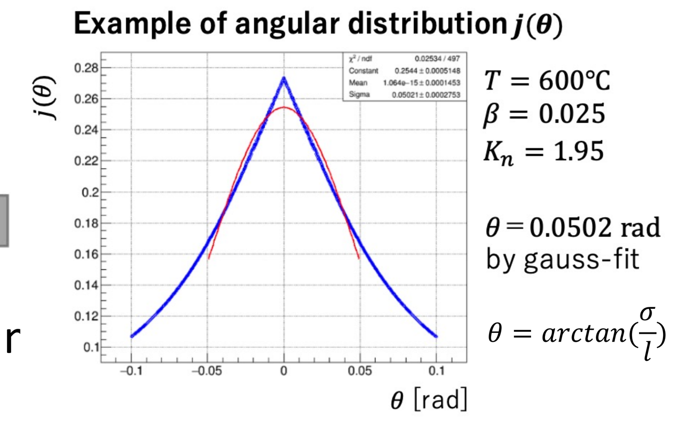
Aim for a sheet-like atomic beam

How to treat the atomic beam

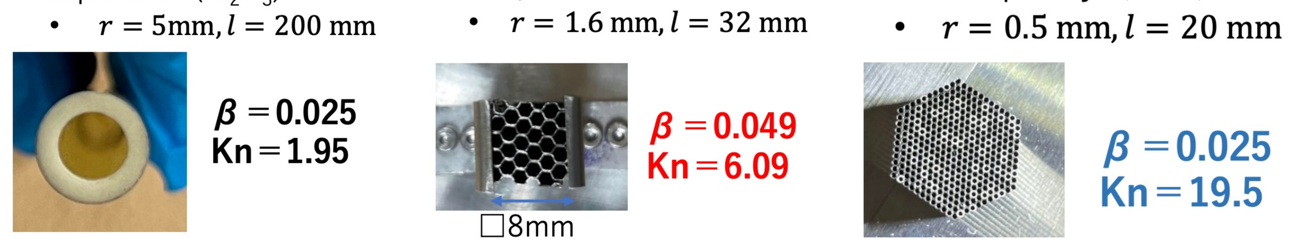
$K_n = \frac{\lambda}{D}$ — Continue $K_n \sim 0$; Intermediate $(0.01 < K_n < 1)$; Molecular flow $K_n > 1$

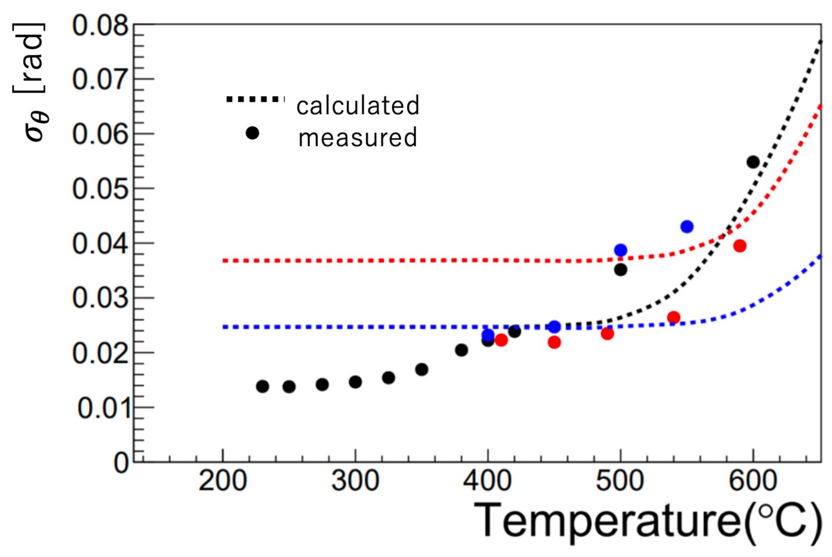
$\beta = \frac{2r}{L}$ — $2r$: diameter; L : length

Example of angular distribution $j(\theta)$

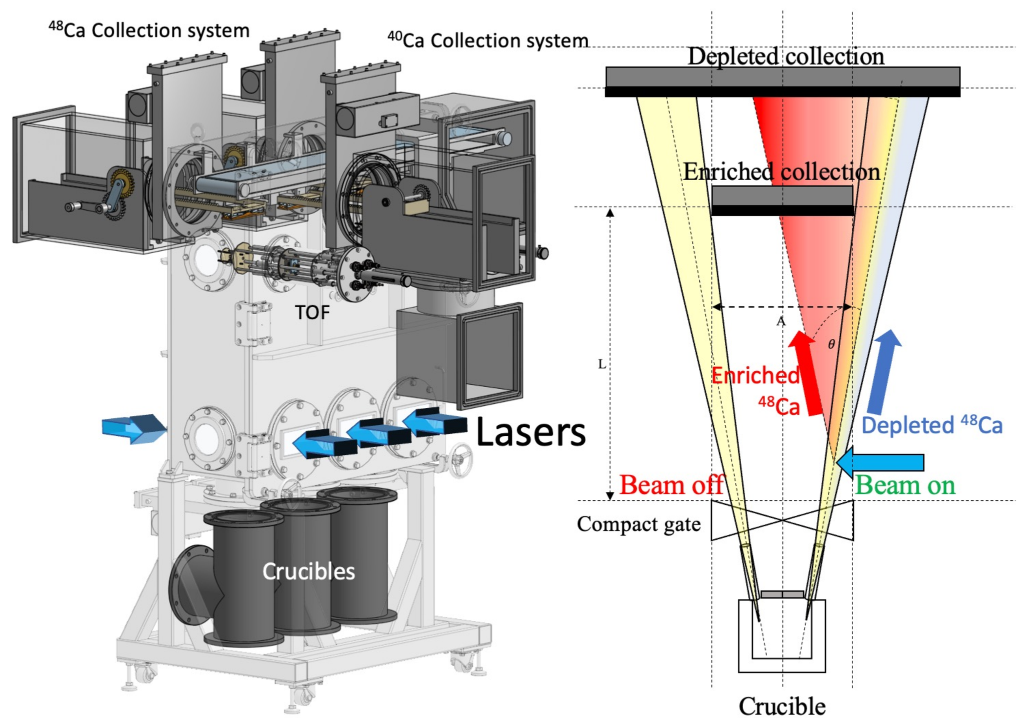


Simple tube (Al_2O_3)	Honeycomb nozzle (SUS)	Micro capillary (SUS)
r = 5mm, l = 200 mm	r = 1.6 mm, l = 32 mm	r = 0.5 mm, l = 20 mm
β = 0.025, Kn = 1.95	β = 0.049, Kn = 6.09	β = 0.025, Kn = 19.5





The spread angle of the Ca atomic beam measured by TOF

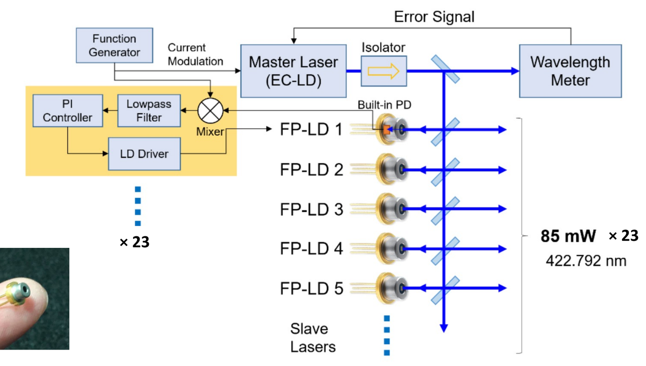


*Current design aims to get up to 2 mol/year production rate of ^{48}Ca

3. Laser

Laser requirements

- ✓ Wavelength: ~422.792 nm
- ✓ Frequency stability: <2 MHz rms
- ✓ Power scalability: >100 W (1 unit)
- ✓ Long life time: >30,000 hours
- ✓ Continuous wave (CW)
- ✓ High efficiency
- ✓ Low cost

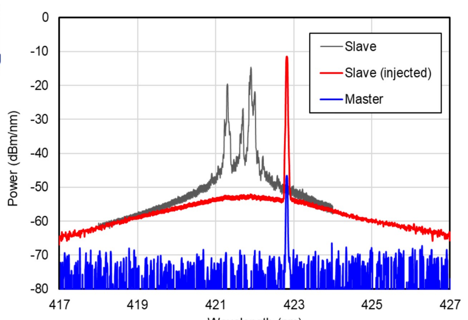


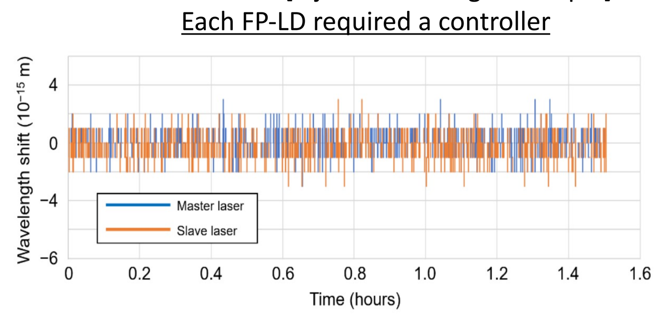
EC-LD + FP-LD [injection locking technique]
Each FP-LD required a controller

Kyoto University Institute for Chemical Research

Institute of Laser Engineering Osaka University

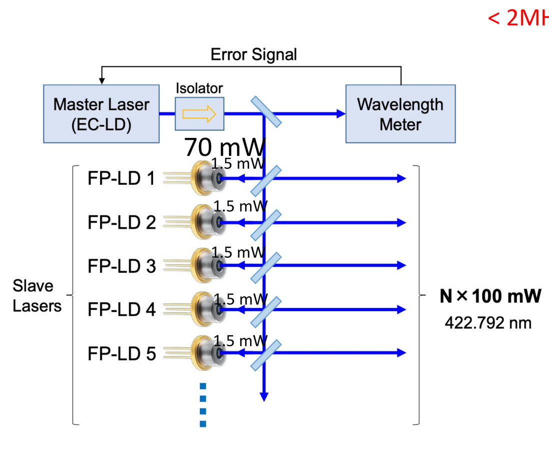
Single frequency was obtained.



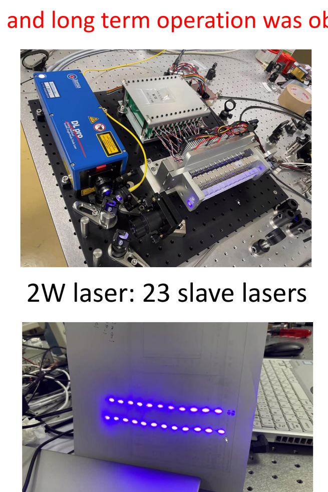


2 MHz rms → 422.792xxxxx ±0.0000006 nm

< 2MHz rms and long term operation was obtained



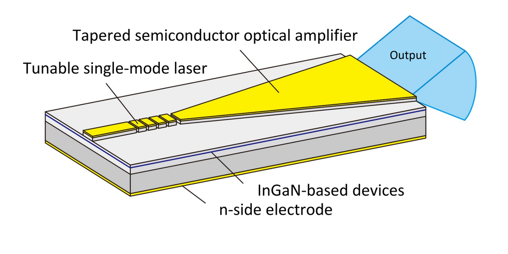
Power scalability of FP-LD laser system



2W laser: 23 slave lasers

Development of tapered semiconductor optical amplifier (SOA)

Collaborator: M. Uemukai (Osaka Univ.)

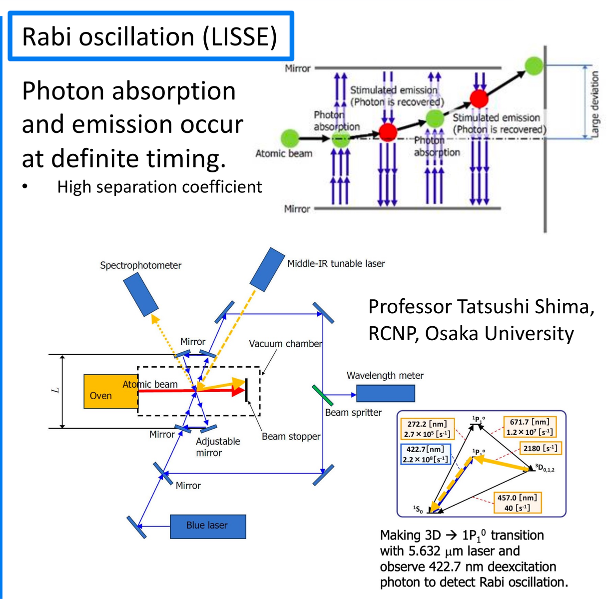


Integrated single-frequency master laser and power amplifier system.

MOPA laser - Development of tapered semiconductor optical amplifier (SOA)
Professor Masahiro Uemukai,
Osaka University

Rabi oscillation (LISSE)

Photon absorption and emission occur at definite timing.
- High separation coefficient



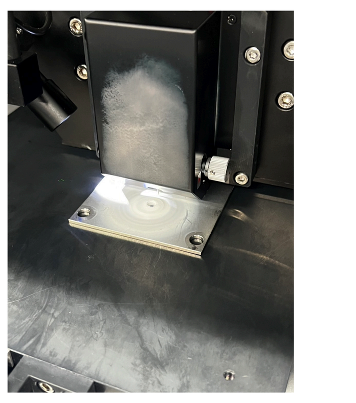
Professor Tatsushi Shima,
RCNP, Osaka University

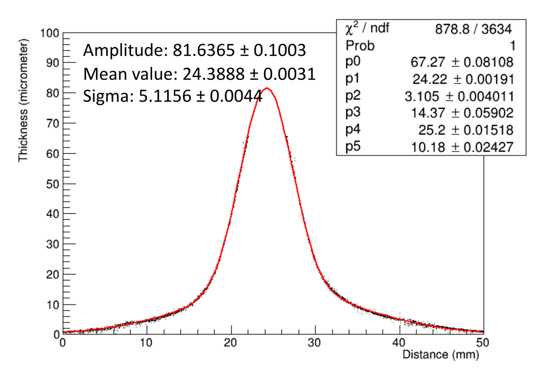
Making 3D → 1P1 transition with 5.632 μm laser and observe 422.7 nm deexcitation photon to detect Rabi oscillation.

4. Collection

RCNP

Determine the collection efficiency on substrate material

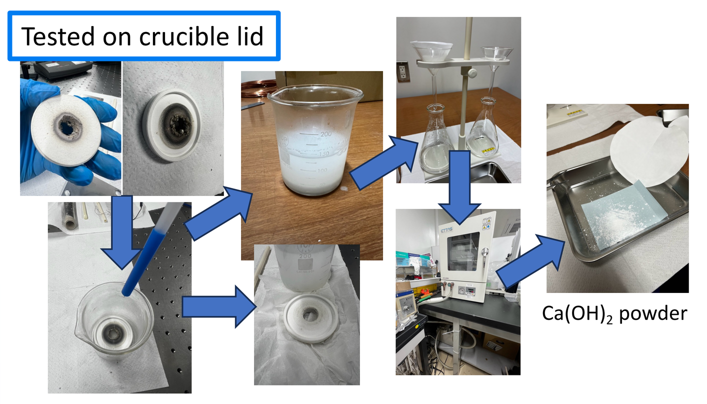




- Thickness measurement was performed using the stylus method @Wakasa-wan.
  - Consider an appropriate substrate material

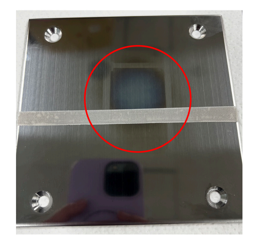
Calcium recovery by chemical method: production of $Ca(OH)_2$

Tested on crucible lid



$Ca + 2H_2O \rightarrow 2Ca(OH)_2 + H_2$
Expose to air: $2Ca + O_2 \rightarrow 2CaO$
$CaO + CO_2 \rightarrow 2CaCO_3$
Reacte with DDW $CaO + H_2O \rightarrow Ca(OH)_2$
Sampling for ICP-MS measurement:
$Ca(OH)_2 + 2HCl \rightarrow CaCl_2 + 2H_2O$
$CaCO_3 + 2HCl \rightarrow CaCl_2 + CO_2 + H2O$

Consolidated substrate



Expose to air to form CaO

↓

Soak with water in an ultrasonic cleaner to get $Ca(OH)_2$

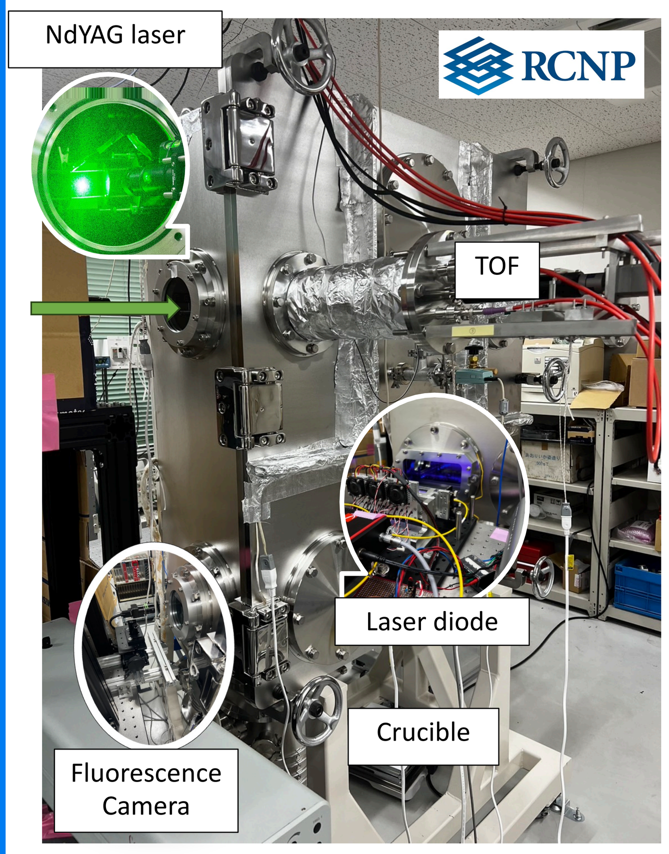
↓

Settling and filtration of the powder and drying it in an oven heater

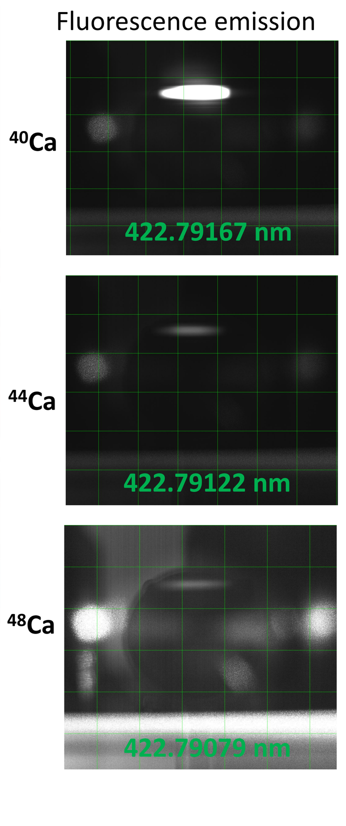
↓

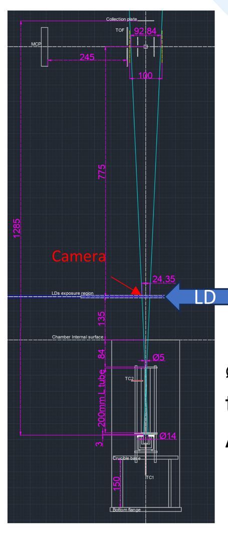
Sampling and dissolving it in HCl to get $CaCl_2$ (aq) and measure the isotope ratio by ICP-MS

5. Production



Fluorescence emission



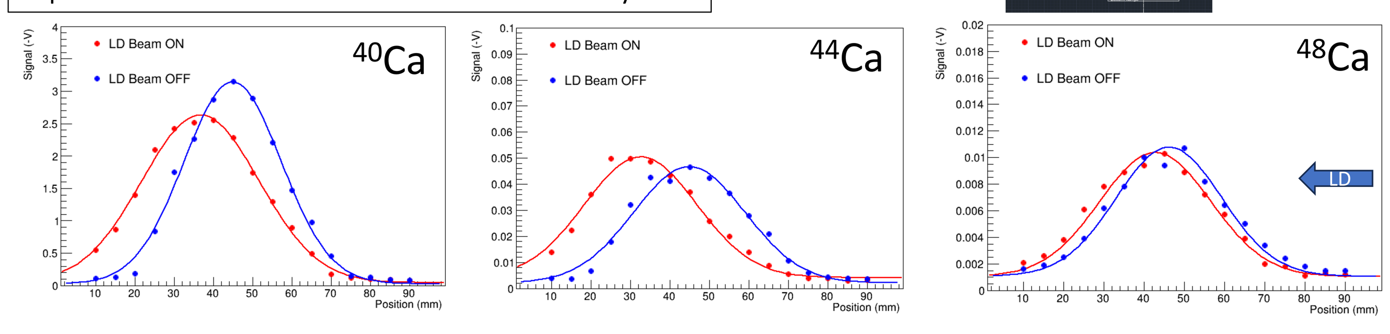


Schematic diagram of the system

single

⌀5mm, L=200m tube collimator Al_2O_3

Spatial distribution of Ca atomic beam measured by TOF



Displacement = 8.20±0.38 mm | Displacement = 11.34±0.77 mm | Displacement = 3.40±0.72 mm

What is the required laser power?

Photon energy: 4.7×10^{-19} J @ 423 nm
Number of photons absorbed by 1 atom: 1,000
→ Number of ^{48}Ca produced by 1 W laser: 2×10^{15} sec^{-1} → ~5 g/W/year

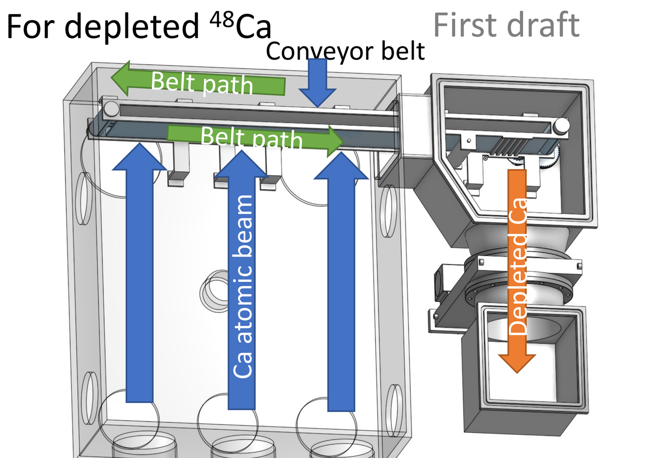
>200 W of laser power produces ^{48}Ca of 1 kg/year.

Current	soon	Near future	Future
100 mW →	2 W →	2 kW →	60 kW
	(~10 g / year)	(~10 kg / year)	(~300 kg / year)

- Automated production system
  ~42.8 kg of ^{nat}Ca -> 2 mol of ^{48}Ca

For depleted ^{48}Ca

First draft



Depleted ^{48}Ca deposition box